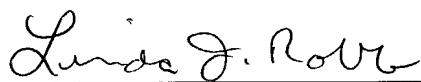


FORM PTO-1390 (REV 10-00)		U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		KRO0105PUSA		
INTERNATIONAL APPLICATION NO PCT/US00/41086		INTERNATIONAL FILING DATE 05 October 2000 (05 10 00)	U S APPLICATION NO (If known, see 37 C F R 1.5) 10/089960	
TITLE OF INVENTION UV CURABLE COMPOSITIONS FOR PRODUCING ELECTROLUMINESCENT COATINGS		PRIORITY DATE CLAIMED 06 October 1999 (06.10.99)		
APPLICANT(S) FOR DO/EO/US Roy C. Krohn				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
1. <input checked="" type="checkbox"/>	This is a FIRST submission of items concerning a filing under 35 U S C 371.			
2. <input type="checkbox"/>	This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U S.C. 371.			
3. <input type="checkbox"/>	This express request to begin national examination procedures (35 U S C 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).			
4. <input checked="" type="checkbox"/>	A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.			
5. <input checked="" type="checkbox"/>	A copy of the International Application as filed (35 U.S.C. 371(c)(2)) a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau) b. <input type="checkbox"/> has been transmitted by the International Bureau. c. <input checked="" type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)			
6. <input type="checkbox"/>	A translation of the International Application into English (35 U S.C. 371(c)(2))			
7. <input type="checkbox"/>	Amendments to the claims of the International Application Under PCT Article 19 (35 U S C 371(c)(3)) a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau) b. <input type="checkbox"/> have been transmitted by the International Bureau c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made.			
8. <input type="checkbox"/>	A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).			
9. <input checked="" type="checkbox"/>	An oath or declaration of the inventor(s) (35 U S C 371(c)(4))			
10. <input type="checkbox"/>	A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U S.C. 371(c)(5))			
Items 11. to 16. below concern document(s) or information included:				
11. <input type="checkbox"/>	An Information Disclosure Statement under 37 CFR 1.97 and 1.98.			
12. <input checked="" type="checkbox"/>	An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included			
13. <input type="checkbox"/>	A FIRST preliminary amendment <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment			
14. <input type="checkbox"/>	A substitute specification.			
15. <input type="checkbox"/>	A change of power of attorney and/or address letter			
16. <input type="checkbox"/>	Other items or information:			

"Express Mail" Mailing Label No.: EV 011 677 804 US

Date of Deposit: April 4, 2002

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" under 37 C.F.R. 1.10 on the date indicated above and is addressed to: P.O. Box PCT, Commissioner for Patents, United States Patent and Trademark Office, Washington, D.C. 20231



Linda J. Robb

U.S. APPLICATION NO. (If known, see 37 C F R 1.5) 101089960	INTERNATIONAL APPLICATION NO PCT/US00/41086	ATTORNEY'S DOCKET NUMBER KRO0105PCT
17. <input checked="" type="checkbox"/> The following fees are submitted.	CALCULATIONS PRO USE ONLY	
BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1,040.00		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00		
ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 890.00		
Surcharge of \$130.00 for furnishing the oath or declaration later than <u>20</u> <u>30</u> months from the earliest claimed priority date (37 CFR 1.492(e)).		
\$ 00.00		
CLAIMS	NUMBER FILED	NUMBER EXTRA
Total claims	25 - 20 = 5	X \$18.00
Independent claims	5 - 3 = 2	X \$84.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)		+ \$280.00
TOTAL OF ABOVE CALCULATIONS = \$ 1,148.00		
Reduction by ½ for filing by small entity. Applicant claims small entity status		
\$ 00.00		
SUBTOTAL = \$ 1,148.00		
Processing fee of \$130.00 for furnishing the English translation later than <u>20</u> <u>30</u> months from the earliest claimed priority date (37 CFR 1.492(f))		
+ \$ 00.00		
TOTAL NATIONAL FEE = \$ 1,148.00		
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		
+ \$ 40.00		
TOTAL FEES ENCLOSED = \$ 1,188.00		
Amount to be: refunded \$ charged \$		
a. <input checked="" type="checkbox"/> A check in the amount of <u>\$ 1,188.00</u> to cover the above fees is enclosed		
b. <input type="checkbox"/> Please charge my Deposit Account No. <u>02-3978</u> in the amount of \$ _____ to cover the above fees A duplicate copy of this sheet is enclosed		
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>02-3978</u> . A duplicate copy of this sheet is enclosed.		
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.		
SEND ALL CORRESPONDENCE TO:		
Mr. James W. Proscia BROOKS & KUSHMAN P.C. 1000 TOWN CENTER, 22 ND FLOOR SOUTHFIELD, MI 48075 PHONE: (248) 358-4400 FAX: (248) 358-3351		
Signature: <u>James Proscia</u>		
Name: <u>JAMES W PROSCIA</u>		
Registration No.: <u>47,010</u>		

UV CURABLE COMPOSITIONS FOR PRODUCING ELECTROLUMINESCENT COATINGS

TECHNICAL FIELD

5 The present invention relates to the active layer in an electroluminescent device.

BACKGROUND ART

10 A typical electroluminescent device is a multilayer thin film structure that emits visible light when activated by an applied voltage. The active layer in such a device will contain a phosphor. This active layer has previously been deposited applying various curable compositions to a suitable substrate followed by ultraviolet (UV) light curing or heat curing. The usual compositions, however, contain organic solvents that do not incorporate into the active layer after curing. Such solvent based systems are undesirable because of the hazards and expenses associated with volatile organic solvents.

15 UV radiation curable compositions are applied to a substrate through spraying, screen printing, dipping or brushing for the protection or decoration of the substrate. In the usual application, a substrate such as metals, glass, or plastics is coated with the composition and then UV light is introduced to complete the curing process. The UV curable compositions offer many advantages over typical heat 20 curable compositions.

25 Heat curable compositions require the use of organic solvents that contain a significant amount of volatile organic compounds (VOCs). These VOCs escape into the atmosphere while the heat curable composition dries. Such solvent based systems are undesirable because of the hazards and expenses associated with VOCs. The hazards include water and air pollution and the expenses include the cost of complying with strict government regulation on solvent emission levels. In

contrast, UV curable compositions contain reactive monomers instead of solvents; thus eliminating the detrimental effects of the VOCs.

The use of heat curable compositions not only raises environmental concerns but other disadvantages exist with their use as well. Heat curable compositions suffer from slow cure times which lead to decreased productivity. These compositions require high energy for curing due to energy loss as well as the energy required to heat the substrate. Additionally, many heat curable compositions yield poor film properties that result in decreased value of the end product.

In a typical electroluminescent device, the active layer comprises one layer of a multilayer electroluminescent device. An example of such a device would contain a substrate made of polycarbonate or glass coated with a transparent conductor such as fluorine doped tin oxide. Metallic grid lines are patterned onto the substrate. The active layer is then applied by screen printing the electroluminescent composition onto the substrate with gridlines. A dielectric coating is then optionally applied over the structure. Finally, the device is coated with a metallic backing. The active layer is such electroluminescent devices typically contains a phosphor. Such phosphor may or may not be encapsulated with various oxides or nitrides. Encapsulation protects the phosphor from the deleterious environmental effects.

20

DISCLOSURE OF INVENTION

It is an object of the present invention to provide an improved electroluminescent composition that is curable by ultraviolet light.

25

It is another object of the present invention to provide an improved electroluminescent composition that can be applied by spraying, screen printing, dipping, and brushing.

It is still another object of the present invention to provide an improved electroluminescent composition that comprises either an encapsulated or an unencapsulated phosphor.

It is yet another object of the present invention to provide an improved 5 electroluminescent composition that comprises at least one aliphatic acrylated oligomer that can be used to coat a substrate such that no significant amount of volatile organic solvents do not become incorporated in the coating after the composition is cured.

The present invention discloses an ultraviolet light curable 10 electroluminescent composition and method for making such a composition that may be used to produce an electroluminescent active layer. In this context, an active layer is a layer that when incorporated in a suitable device emits light when a voltage is applied. The disclosed composition does not contain any significant amount of volatile organic solvents that do not become incorporated in the active layer after 15 curing. Specifically, the electroluminescent composition contains 5% or less volatile organic solvents by weight. It is an advantage of the present invention that the deposition of the dielectric layer in such a device is optional when the disclosed electroluminescent composition is used to deposit the active layer.

In accordance with one aspect of the invention, an ultraviolet light 20 curable electroluminescent composition is provided. The electroluminescent composition comprises a mixture of one or more aliphatic acrylated oligomers, wherein the aliphatic acrylated oligomer mixture is present in an amount of about 10% to 40% of the electroluminescent composition. All percentages of the electroluminescent composition as expressed in this document refer to the weight 25 percentage of the stated component to the total mass of the electroluminescent composition.

The electroluminescent composition preferably comprises an isobornyl acrylate monomer in an amount of about 4% to 30% of the electroluminescent composition, optionally an adhesion promoter in an amount of

1% to 10%, a photoinitiator in an amount of about 0.5% to 6% of the electroluminescent composition, optionally, a flow promoting agent in an amount of about 0.1% to 5% of the electroluminescent composition, and an electroluminescent phosphor in an amount of 28% to 80%. The electroluminescent phosphor may either be encapsulated or unencapsulated.

In accordance with yet another aspect of the invention, a method is provided for depositing a electroluminescent coating on a substrate. The method comprises a first step of applying to the substrate a electroluminescent-containing fluid-phase composition ("electroluminescent composition"). The electroluminescent composition comprises a mixture of aliphatic acrylated oligomers, wherein the aliphatic acrylated oligomer is present in an amount of about 10% to 40% of the electroluminescent composition. The electroluminescent composition also includes an isobornyl acrylate monomer in an amount of about 4% to 30% of the electroluminescent composition, a photoinitiator in an amount of about 0.5% to 6% of the electroluminescent composition, a flow promoting agent in an amount of about 0.1% to 5% of the electroluminescent composition, a copper activated zinc sulfide electroluminescent phosphor in an amount of 28% to 80%, and optionally an adhesion promoter in an amount of 1% to 10%.

The method also includes a second step of illuminating the electroluminescent composition on the substrate with an ultraviolet light to cause the electroluminescent composition to cure into the electroluminescent coating.

In accordance with this method, the electroluminescent composition can be selectively deposited on the substrate at specific locations where electroluminescent plating is desired. It need not be applied to the entire substrate.

25

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a graph that the light intensity over time of an electroluminescent device incorporating an active layer made with the composition of the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION**Electroluminescent Compositions**

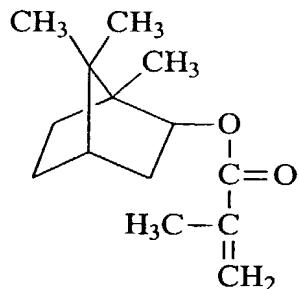
Reference will now be made in detail to presently preferred compositions or embodiments and methods of the invention, which constitute the best modes of practicing the invention presently known to the inventor.

In accordance with one aspect of the invention, a presently preferred ultraviolet light curable electroluminescent composition ("electroluminescent composition") is provided. In this preferred embodiment, the electroluminescent composition includes a mixture of aliphatic acrylated oligomers. The aliphatic acrylated oligomer mixture is preferably present in an amount of about 10% to 40% of the weight of the electroluminescent composition. In a particularly preferred embodiment the aliphatic acrylated oligomer mixture is present in an amount of about 34% of the weight of the electroluminescent composition. In another particularly preferred embodiment the aliphatic acrylated oligomer mixture is present in an amount of about 12% of the weight of the electroluminescent composition.

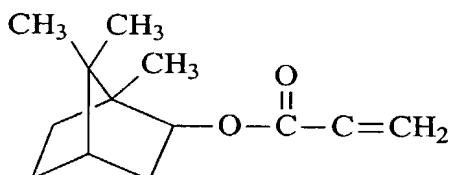
The aliphatic acrylated oligomer preferably comprises one or more urethane oligomers. Suitable aliphatic acrylated oligomers include Radcure Ebecryl 244 (aliphatic urethane diacrylate diluted 10% with 1,6-hexanediol diacrylate), Ebecryl 264 (aliphatic urethane triacrylate diluted 15% with 1,6-hexanediol diacrylate), Ebecryl 284 (aliphatic urethane diacrylate diluted 10% with 1, 6- hexanediol diacrylate) commercially available from Radcure UCB Corp. of Smyrna, Georgia; Sartomer CN-961E75 (aliphatic urethane diacrylate blended with 25% ethoxylated trimethylol propane triacrylate), CN-961H81 (aliphatic urethane diacrylate blended with 19% 2(2-ethoxyethoxy)ethyl acrylate), CN-963A80 (aliphatic urethane diacrylate blended with 20% tripropylene glycol diacrylate), CN-964 (aliphatic urethane diacrylate), CN-966A80 (aliphatic urethane diacrylate blended with 20% tripropylene glycol diacrylate), CN-982A75 (aliphatic urethane diacrylate blended with 25% tripropylene glycol diacrylate) and CN-983 (aliphatic urethane diacrylate), commercially available from Sartomer Corp. of Exton, Pennsylvania; TAB FAIRAD 8010, 8179, 8205, 8210, 8216, 8264, M-E-15, UVU-316, commercially available

from TAB Chemicals of Chicago, Illinois; and Echo Resin ALU-303, commercially available from Echo Resins of Versaille, Missouri; and Genomer 4652, commercially available from Rahn Radiation Curing of Aurora, IL. The preferred aliphatic acrylated oligomers include Ebecryl 264 and Ebecryl 284. Ebecryl 264 is an aliphatic urethane triacrylate of 1200 molecular weight supplied as an 85% solution in hexanediol diacrylate. Ebecryl 284 is aliphatic urethane diacrylate of 1200 molecular weight diluted 10% with 1,6-hexanediol diacrylate. Combinations of these materials may also be employed herein.

The preferred electroluminescent composition also includes an isobornyl acrylate monomer preferably in an amount of about 4% to 30% of the electroluminescent composition. In one particularly preferred embodiment of the present invention, the isobornyl acrylate monomer is present in an amount of about 20% of the electroluminescent composition. In another particularly preferred embodiment of the present invention, the isobornyl acrylate monomer is present in an amount of about 8% of the electroluminescent composition. Suitable isobornyl acrylate monomers include Sartomer SR423 (isobornyl methacrylate):



and SR506 (isobornyl acrylate):



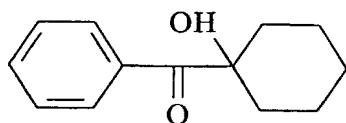
available from Sartomer Corp.; Radcure IBOA (isobornyl acrylate), commercially available from Radcure Corp.; IBOA and IBOMA, commercially available from CPS Chemical of Bradford, England; and Genomer 1121, commercially available from Rahn Radiation Curing. The preferred isobornyl acrylate monomer is Radcure 5 IBOA, commercially available from Radcure Corp. Radcure IBOA is a high purity, low color monomer. Combinations of these materials may also be employed herein.

The preferred electroluminescent composition may also include an adhesion promoter preferably in an amount of about 1% to 10% the electroluminescent composition. In one particularly preferred embodiment of the 10 present invention the adhesion promoter is present in an amount of about 7%. In another particularly preferred embodiment of the present invention the adhesion promoter is present in an amount of about 3% of the electroluminescent composition. Suitable adhesion promoters include Ebecryl 168, commercially available from Radcure Corp.; and Sartomer CN 704 (acrylated polyester adhesion promoter) and 15 CD 9052 (trifunctional acid ester), commercially available from Sartomer Corp. The preferred adhesion promoter is Ebecryl 168 which is a methacrylated acidic adhesion promoter. Combinations of these materials may also be employed herein.

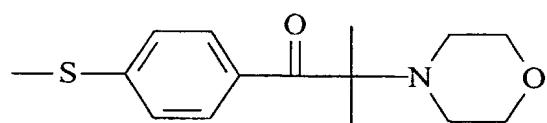
The preferred electroluminescent composition also includes an electroluminescent phosphor. Preferably the electroluminescent phosphor is a 20 copper activated zinc sulfide electroluminescent phosphor. The copper activated zinc sulfide electroluminescent phosphor is preferably present in an amount of about 28% to 80% of the electroluminescent composition. The copper activated zinc sulfide electroluminescent phosphor may either be encapsulated or unencapsulated. In one 25 particularly preferred embodiment of the present composition the zinc sulfide electroluminescent phosphor is present in an amount of about 33%. In another particularly preferred embodiment of the present invention, the electroluminescent phosphor is a mixture of an encapsulated and unencapsulated phosphor present in a total amount of about 80% of the weight of the electroluminescent composition 30 wherein the encapsulated phosphor is about 50% of the electroluminescent composition and the unencapsulated phosphor is about 25% of the electroluminescent composition. Suitable encapsulated electroluminescent phosphors include TNE 100,

TNE 120, TNE 200 TNE 210, TNE 220, TNE 230, TNE 300, TNE 310, TNE 320, TNE 400, TNE 410 TNE 420, TNE 430, TNE 500, TNE 510, TNE 520, TNE 600, TNE 620, TNE 700, TNE 720, NE 100, NE 120, NE 200, NE 210, NE 220, NE 230, NE 300, NE 310, NE 320, NE 400, NE 410 NE 420, NE 430, NE 500, NE 510, NE 520, NE 600, NE 620, NE 700, NE 720, ANE 200, ANE 230, AND 400, and ANE 430 commercially available from Osram Sylvania. Suitable unencapsulated electroluminescent phosphors include 723 EL, 727 EL, 728 EL, 729 EL, 813 EL, and 814 EL commercially available from Osram Sylvania. These materials may emit red, green, yellow, blue or orange colored light based upon the particular phosphor employed. The preferred electroluminescent phosphors are TNE 100, TNE 200, TNE 410, and TNE 700, commercially available from Osram Sylvania.

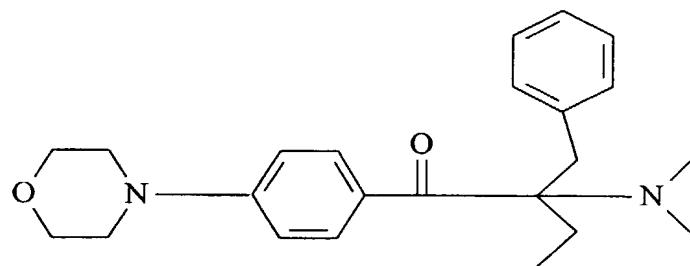
This preferred electroluminescent composition also includes a photoinitiator preferably in an amount of about 0.5% to 6% of the electroluminescent composition. In one particularly preferred embodiment of the present invention the photoinitiator is present in an amount of about 3%. In another particularly preferred embodiment of the present invention the photoinitiator is present in an amount of about 1% of the electroluminescent composition. Suitable photoinitiators include Irgacure 184 (1-hydroxycyclohexyl phenyl ketone),



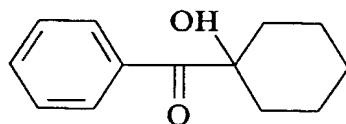
Irgacure 907 (2-methyl-1-[4-(methylthio)phenyl]-2-morpholino propan-1-one),



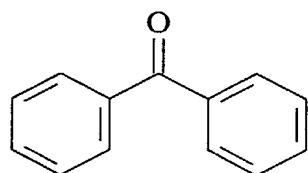
20 Irgacure 369 (2-benzyl-2-N,N-dimethylamino-1-(4-morpholinophenyl)-1-butanone),



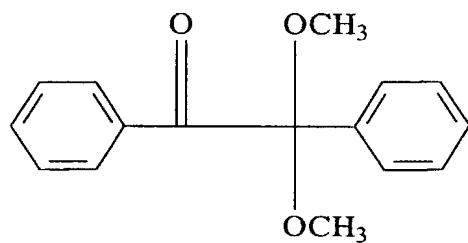
Irgacure 500 (the combination of 50% 1-hydroxy cyclohexyl phenyl ketone,



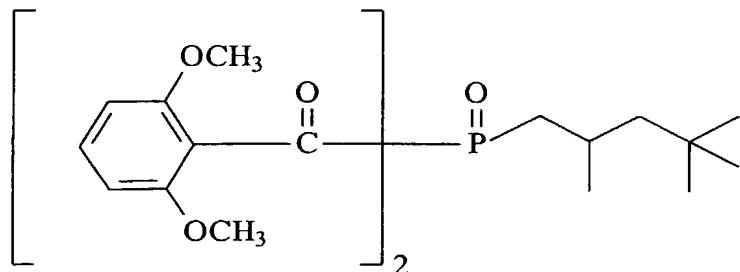
and 50% benzophenone),



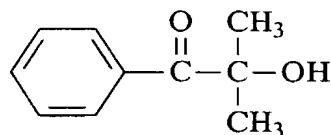
Irgacure 651 (2,2-dimethoxy-1,2-diphenylethan-1-one),



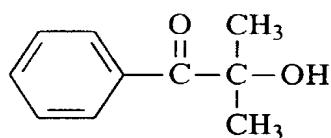
Irgacure 1700 (the combination of 25% bis(2,6-dimethoxybenzoyl-2,4-,4-trimethyl pentyl) phosphine oxide,



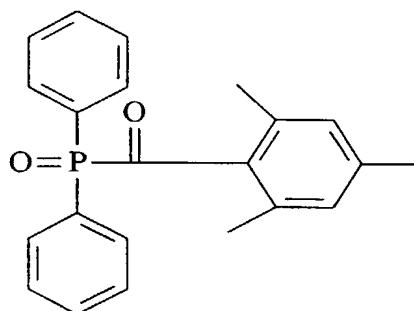
and 75% 2-hydroxy-2-methyl-1-phenyl-propan-1-one),



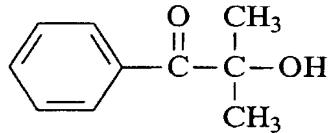
and DAROCUR 1173 (2-hydroxy-2-methyl-1phenyl-1-propane),



5 and DAROCUR 4265 (the combination of 50% 2,4,6- trimethylbenzoyldiphenyl- phosphine oxide,



and 50% 2-hydroxy 2-methyl-1-phenyl-propan-1-one),



available commercially from Ciba-Geigy Corp., Tarrytown, N.Y.; CYRACURE UVI-6974 (mixed triaryl sulfonium hexafluoroantimonate salts) and cyracure UVI-6990 (mixed triaryl sulfonium hexafluorophosphate salts) available commercially from Union Carbide Chemicals and Plastics Co. Inc., Danbury, Connecticut; and Genocure CQ, Genocure BOK, and Genocure M.F., commercially available from Rahn Radiation Curing. The preferred photoinitiator is Irgacure 1700 commercially available from Ciba-Geigy of Tarrytown, New York. Combinations of these materials may also be employed herein.

10 The preferred electroluminescent composition still further includes a flow promoting agent preferably in an amount of about 0.1% to 5% of the electroluminescent composition. In one particularly preferred embodiment of the present invention the flow promoting agent is present in an amount of about 3%. In another particularly preferred embodiment of the present invention the flow 15 promoting agent is present in an amount of about 1% of the electroluminescent composition. Suitable flow promoting agents include Genorad 17, commercially available from Rahn Radiation Curing; and Modaflow, commercially available from Monsanto Chemical Co., St. Louis, Missouri. The preferred flow promoting agent is Modaflow which is an ethyl acrylate and 2-ethylhexyl acrylate copolymer that 20 improves the flow of the composition. Combinations of these materials may also be employed herein.

To illustrate, the following example sets forth a presently preferred electroluminescent composition according to this aspect of the invention.

Example 1

This example provides a preferred electroluminescent composition according to the invention that can be used for deposition on the surface of a substrate such as a glass or polycarbonate substrate. Such a substrate may first be 5 coated with a transparent conductor and silver grid lines. The electroluminescent composition was made from the following components:

	Component	Approximate Mass %
	Ebecryl 264	16.7
10	Ebecryl 284	16.7
	IBOA	20.0
	Ebecryl 168	6.7
	Modaflow	3.3
	Irgacure 1700	3.3
15	TNE 200	33.3
	Total	100.00

In this example the IBOA and Irgacure 1700 are mixed in a pan with a propeller blade mixer for 30 seconds at a speed of 500 to 1000 rpm. In the next step, the Ebecryl 264, the Ebecryl 284, and the Modaflow are introduced into the pan and mixed for 1 to 2 minutes at a speed of 2000 rpm. In the final step, the 20 Ebecryl 168 adhesion promoter and TNE 200 phosphor are introduced into the pan and are mixed for 1 to 2 minutes at a speed of 2000 rpm. The temperature during mixing is monitored. The mixing is temporarily suspended if the temperature exceed 100°F. This particular electroluminescent composition may be used by the method below to produce an active layer that emits bluish-green light.

25

Example 2

This example provides a preferred electroluminescent composition according to the invention that can be used for deposition on the surface of a

substrate such as a glass or polycarbonate substrate. Such a substrate may first be coated with a transparent conductor and silver grid lines. The electroluminescent composition was made from the following components:

	Component	Approximate Mass %
5	Ebecryl 264	6.3
	Ebecryl 284	6.3
	IBOA	7.5
	Ebecryl 168	2.5
	Modaflow	1.3
	Irgacure 1700	1.3
	ANE 430	49.8
	813 EL	25.0
Total		100.00

In this example the IBOA and Irgacure 1700 are mixed in a pan with a propeller blade mixer for 30 seconds at a speed of 500 to 1000 rpm. In the next step, the Ebecryl 264, the Ebecryl 284, and the Modaflow are introduced into the pan and mixed for 1 to 2 minutes at a speed of 2000 rpm. In the final step, the Ebecryl 168 adhesion promoter, ANE 430 phosphor, and 813 EL phosphor are introduced into the pan and are mixed for 1 to 2 minutes at a speed of 2000 rpm. The temperature during mixing is monitored. The mixing is temporarily suspended if the temperature exceed 100°F. This particular electroluminescent composition may be used by the method below to produce an active layer that emits bluish-green light.

Figure 1 is a graph of the change in light output intensity over time for an electroluminescent device incorporating an active layer made from the composition in Example 2. The electroluminescent device is made from a 4 inch by 4 inch glass substrate coated with a transparent conductor. Metallic grid lines are patterned onto the substrate. The active layer is then applied by screen printing the

electroluminescent composition described in Example 2 onto the substrate. Finally, the device is coated with a silver backing. Figure 1 shows that the light intensity of such a device when powered by a 12.5 volt transformer at 100 Hz initially is approximately 20 foot candles per square centimeter and that over a period of 5 approximately two months the light intensity decreases and thereafter stabilizes at a value of approximately 10 foot candles per square centimeter. Furthermore, electroluminescent devices made from the composition in Example 2 have achieved light intensities as high as 88 foot candles per square centimeter for 1 inch by 1 inch devices when powered at 240 watts and 2000 Hz.

10 **Method for Depositing an Electroluminescent Coating on a Substrate**

In accordance with still another aspect of the invention, a method is provided for depositing an electroluminescent coating on a suitable substrate. This method is utilized in depositing the active layer in a multilayer electroluminescent device. The method comprises a first step of applying a phosphor-containing fluid-phase composition ("electroluminescent composition") to the substrate.

20 The electroluminescent composition comprises an aliphatic acrylated oligomer, the aliphatic acrylated oligomer preferably present in an amount of about 10% to 40% of the electroluminescent composition; an isobornyl acrylate monomer preferably present in an amount of about 4% to 30% of the electroluminescent composition; an photoinitiator preferably present in an amount of a preferably present bout 0.5% to 6% of the electroluminescent composition; a flow promoting agent in an amount of about 0.1% to 5% of the electroluminescent composition, and a zinc sulfide containing phosphor in an amount of 28% to 80%. The preferred electroluminescent compositions according to this method are those described 25 herein, for example, including the compositions described in example 1 and example 2.

30 The electroluminescent composition may be applied to the substrate using a number of different techniques. The electroluminescent composition may be applied, for example, by direct brush application, or it may be sprayed onto the substrate surface. It also may be applied using a screen printing technique. In such

screen printing technique, a "screen" as the term is used in the screen printing industry is used to regulate the flow of liquid composition onto the substrate surface. The electroluminescent composition typically would be applied to the screen as the latter contacts the substrate. The electroluminescent composition flows through the 5 silk screen to the substrate, whereupon it adheres to the substrate at the desired film thickness. Screen printing techniques suitable for this purpose include known techniques, but wherein the process is adjusted in ways known to persons of ordinary skill in the art to accommodate the viscosity, flowability, and other properties of the liquid-phase composition, the substrate and its surface properties, etc. Flexographic 10 techniques, for example, using pinch rollers to contact the electroluminescent composition with a rolling substrate, also may be used.

The method includes a second step of illuminating the electroluminescent-containing fluid-phase composition on the substrate with an ultraviolet light to cause the electroluminescent-containing fluid-phase composition 15 to cure into the electroluminescent coating. This illumination may be carried out in any number of ways, provided the ultraviolet light or radiation impinges upon the electroluminescent composition so that the electroluminescent composition is caused to polymerize to form the coating, layer, film, etc., and thereby cures.

Curing preferably takes place by free radical polymerization, which 20 is initiated by an ultraviolet radiation source. The photoinitiator preferably comprises a photoinitiator, as described above.

Various ultraviolet light sources may be used, depending on the application. Preferred ultraviolet radiation sources for a number of applications include known ultraviolet lighting equipment with energy intensity settings of, for 25 example, 125 watts, 200 watts, and 300 watts per square inch.

Additional advantages and modifications will readily occur to those skilled in the art. Therefore, the invention in its broader aspects is not limited to the specific details, representative devices, and illustrative examples shown and

described. Accordingly, departures may be made from such details without departing from the spirit or scope of the general inventive concept.

While embodiments of the invention have been illustrated and described, it is not intended that these embodiments illustrate and describe all possible forms of the invention. Rather, the words used in the specification are words of description rather than limitation, and it is understood that various changes may be made without departing from the spirit and scope of the invention.

WHAT IS CLAIMED IS:

1. An ultraviolet (UV) curable electroluminescent composition comprising:
 - 5 at least one aliphatic acrylated oligomer;
 - an isobornyl acrylate monomer;
 - 10 a photoinitiator; and
 - an electroluminescent phosphor, wherein upon exposure to UV light the electroluminescent composition cures into a layer suitable for use as the active layer in an electroluminescent device and wherein the electroluminescent composition does not contain any significant amount of volatile organic solvents that
 - 15 do not become incorporated in the coating after the electroluminescent composition is cured.
2. The UV curable electroluminescent composition of claim 1, wherein,
 - 15 the aliphatic acrylated oligomer mixture is present in an amount of about 10% to 40% of the weight of the electroluminescent composition;
 - the isobornyl acrylate monomer is present in an amount of about 4% to 30% of the weight of the electroluminescent composition;
 - 20 the photoinitiator is present in an amount of about 0.5% to 6% of the weight of the electroluminescent composition; and
 - the phosphor is present in an amount of about 28% to 80% of the weight of the electroluminescent composition.
3. The UV curable electroluminescent composition of claim 2, wherein the at least one aliphatic acrylated oligomer is at least one urethane oligomer.
4. The UV curable composition of claim 2 further comprising:
 - 25 an adhesion promoter in an amount of about 1% to 10% of the weight of the composition; and

a flow promoting agent in an amount of 0.1 % to 5% of the weight of the electroluminescent composition.

5. The UV curable composition of claim 4 wherein:
the aliphatic acrylated oligomer mixture is present in an amount of
about 34 % of the weight of the electroluminescent composition;
the isobornyl acrylate monomer is present in an amount of about 20%
of the weight of the electroluminescent composition;
the photoinitiator is present in an amount of about 3 % of the weight
of the electroluminescent composition;
10 the phosphor is present in an amount of about 33 % of the weight of
the electroluminescent composition;
the adhesion promoter in an amount of about 7 % of the weight of the
composition; and
the flow promoting agent in an amount of 3 % of the weight of the
15 electroluminescent composition.

6. The UV curable composition of claim 4 wherein:
the aliphatic acrylated oligomer mixture is present in an amount of
about 12 % of the weight of the electroluminescent composition;
20 the isobornyl acrylate monomer is present in an amount of about 8 %
of the weight of the electroluminescent composition;
the photoinitiator is present in an amount of about 1 % of the weight
of the electroluminescent composition;
the phosphor is present in an amount of about 75 % of the weight of
25 the electroluminescent composition;
the adhesion promoter in an amount of about 3 % of the weight of the
composition; and
the flow promoting agent in an amount of 1% of the weight of the
electroluminescent composition.

30 7. The electroluminescent composition of claim 1 wherein the
aliphatic acrylated oligomer in the mixture is selected from the group consisting of:

- a) aliphatic urethane diacrylate diluted 10% by weight with 1,6-hexanediol diacrylate;
- b) aliphatic urethane triacrylate diluted 15% by weight with 1,6-hexanediol diacrylate;
- 5 c) aliphatic urethane diacrylate blended with 20% by weight tripropylene glycol diacrylate;
- d) aliphatic urethane diacrylate blended with 25% by weight ethoxylated trimethylol propane triacrylate;
- 10 e) aliphatic urethane diacrylate blended with 19% by weight 2(2-ethoxyethoxy)ethyl acrylate;
- f) aliphatic urethane diacrylate blended with 20% by weight tripropylene glycol diacrylate;
- 15 g) aliphatic urethane diacrylate blended with 20% by weight tripropylene glycol diacrylate;
- h) aliphatic urethane diacrylate blended with 25% by weight tripropylene glycol diacrylate;
- i) aliphatic urethane diacrylate; and
- j) mixtures thereof.

20 8. The electroluminescent composition of claim 1 wherein the isobornyl acrylate monomer in the mixture is selected from the group consisting of isobornyl acrylate, isobornyl methacrylate, and mixtures thereof.

25 9. The electroluminescent composition of claim 1 wherein the photoinitiator is selected from the group consisting of:

- 1-hydroxycyclohexyl phenyl ketone;
- 2-methyl-1-[4-(methylthio)phenyl]-2-morpholino propan-1-one;
- the combination of 50% 1-hydroxy cyclohexyl phenyl ketone and 50% benzophenone;

30

- 2,2-dimethoxy-1,2-diphenylethan-1-one;
- the combination of 25% bis(2,6-dimethoxybenzoyl-2,4-, 4-trimethyl pentyl phosphine oxide and 75% 2-hydroxy-2-methyl-1-phenyl-propan-1-one;
- 2-hydroxy-2-methyl-1-phenyl-1-propane;

5

the combination of 50% 2,4,6-trimethylbenzoyldiphenyl-phosphine oxide and 50% 2-hydroxy 2-methyl-1-phenyl-propan-1-one;
mixed triaryl sulfonium hexafluoroantimonate salts;
mixed triaryl sulfonium hexafluorophosphate salts; and
mixtures thereof.

10

10. A method for coating a substrate with an electroluminescent composition, the method comprising:

applying the electroluminescent composition to the substrate, wherein the electroluminescent composition includes:

15

an aliphatic acrylated oligomer mixture in an amount of about 10% to 40 % of the weight of the composition;

an isobornyl acrylate monomer in an amount of about 4 % to 30% of the weight of the composition;

an photoinitiator in an amount of about 0.5% to 6% of the weight of the composition;

an adhesion promoter in an amount of about 1% to 10% of the weight of the composition;

a flow promoting agent in an amount of 0.1 % to 5% of the weight of the electroluminescent composition; and

20

an electroluminescent phosphor in an amount of about 28% to 80% of the weight of the composition; and

illuminating the electroluminescent composition on the substrate with an UV light sufficient to cause the electroluminescent composition to cure into a layer suitable for use as the active layer of an electroluminescent device.

25

11. The method of claim 10, wherein the UV light used in illuminating impinges upon the electroluminescent composition so that the electroluminescent composition is caused to form the coating it cures.

30

12. The method of claim 10, wherein the method of applying the electroluminescent composition is spraying.

13. The method of claim 10, wherein the method of applying the electroluminescent composition is screen-printing.

14. The method of claim 10, wherein the method of applying the electroluminescent composition is dipping the substrate into the composition sufficiently to cause the composition to uniformly coat the substrate.

5 15. The method of claim 10, wherein the method of applying the electroluminescent composition is brushing.

10 16. The method of claim 10, wherein the method of applying the electroluminescent composition is selectively depositing to the substrate at predetermined locations.

15 17. A method of a substrate with an ultraviolet (UV) curable electroluminescent composition to form a electroluminiscent coating comprising: applying the electroluminescent composition to the substrate, wherein the composition includes:

20 an aliphatic acrylated oligomer mixture in an amount of about 10% to 40% of the weight of the electroluminiscent composition;

25 an isobornyl acrylate monomer in an amount of about 4% to 30% of the weight of the electroluminescent composition;

a photoinitiator in an amount of about 0.5% to 6% of the weight of the electroluminescent composition;

a flow promoting agent in an amount of 0.1% to 5% of the weight of the electroluminescent composition; and

30 an electroluminescent phosphor in an amount of about 28% to 80% of the weight of the electroluminescent composition; and

35 illuminating the electroluminescent composition on the substrate with an UV light sufficient to cause the electroluminescent composition to cure into a layer suitable for use as the active layer of an electroluminescent device.

18. The method of claim 17, wherein,
the aliphatic acrylated oligomer mixture is about 34% of the weight
of the electroluminescent composition;

5 the isobornyl acrylate monomer is about 20% of the weight of the
electroluminescent composition;

the photoinitiator is about 3% of the weight of the electroluminescent
composition;

10 the flow promoting agent is about 3% of the weight of the
electroluminescent composition; and

the phosphor is about 33% of the weight of the electroluminescent
composition.

19. The method of claim 17, wherein,
the aliphatic acrylated oligomer mixture is about 12% of the weight
of the electroluminescent composition;

15 the isobornyl acrylate monomer is about 8% of the weight of the
electroluminescent composition;

the photoinitiator is about 1% of the weight of the electroluminescent
composition;

20 the flow promoting agent is about 1% of the weight of the
electroluminescent composition; and

the phosphor is about 75% of the weight of the electroluminescent
composition.

25 20. A method of preparing an ultraviolet (UV) curable
electroluminiscent composition comprising:
combining an isobornyl acrylate monomer and a photoinitiator in a
pan to form a first combination;

mixing the first combination;

combining an aliphatic acrylated oligomer mixture and flow
promoting agent with the first mixture to form a second combination;

30 mixing the second combination;

combining an electroluminescent phosphor and an adhesion promoter with the second mixture to form a third combination; and
mixing the third combination.

5 21. A UV curable electroluminescent composition of claim 1,
wherein the electroluminescent phosphor comprises a sulfide electroluminescent phosphor.

22. A UV curable electroluminescent composition of claim 21,
wherein the electroluminescent phosphor comprises a zinc sulfide electroluminescent phosphor.

10 23. A UV curable electroluminescent composition of claim 22,
wherein the electroluminescent phosphor comprises a copper activated zinc sulfide electroluminescent phosphor.

15 24. A UV curable electroluminescent composition of claim 1,
wherein the electroluminescent phosphor is present in an amount between about 28% to 80% of the weight of the electroluminescent composition.

20 25. An ultraviolet (UV) curable electroluminescent composition comprising:
at least one aliphatic acrylated oligomer;
an isobornyl acrylate monomer;
a photoinitiator; and
a phosphor, wherein upon exposure to UV light the electroluminescent composition cures into a layer suitable for use as the active layer in an electroluminescent device and wherein the electroluminescent composition does not contain any significant amount of volatile organic solvents that do not become incorporated in the coating after the electroluminescent composition is cured; and
the phosphor is present in an amount of about 28% to 80% of the weight of the electroluminescent composition.

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
12 April 2001 (12.04.2001)

PCT

(10) International Publication Number
WO 01/25342 A1

(51) International Patent Classification⁷: C09D 4/00, C09K 11/02, H05B 33/20

(21) International Application Number: PCT/US00/41086

(22) International Filing Date: 5 October 2000 (05.10.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
60/157,944 6 October 1999 (06.10.1999) US

(71) Applicant (for all designated States except US): UV SPECIALTIES, INC. [US/US]; 48 N. Airport Drive, Kimball, MI 48074 (US).

(72) Inventor; and

(75) Inventor/Applicant (for US only): KROHN, Roy, C. [US/US]; 3540 Orvall Drive, Fort Gratiot, MI 48059 (US).

(74) Agents: PROSCIA, James, W. et al.; Brooks & Kushman, Twenty-Second Floor, 1000 Town Center, Southfield, MI 48075 (US).

(81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

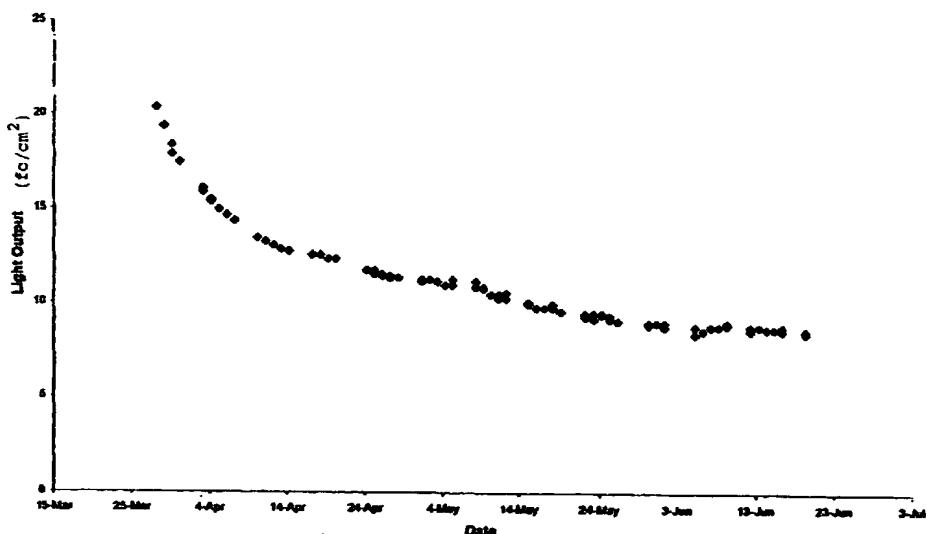
(84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

- With international search report.
- Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: UV CURABLE COMPOSITIONS FOR PRODUCING ELECTROLUMINESCENT COATINGS



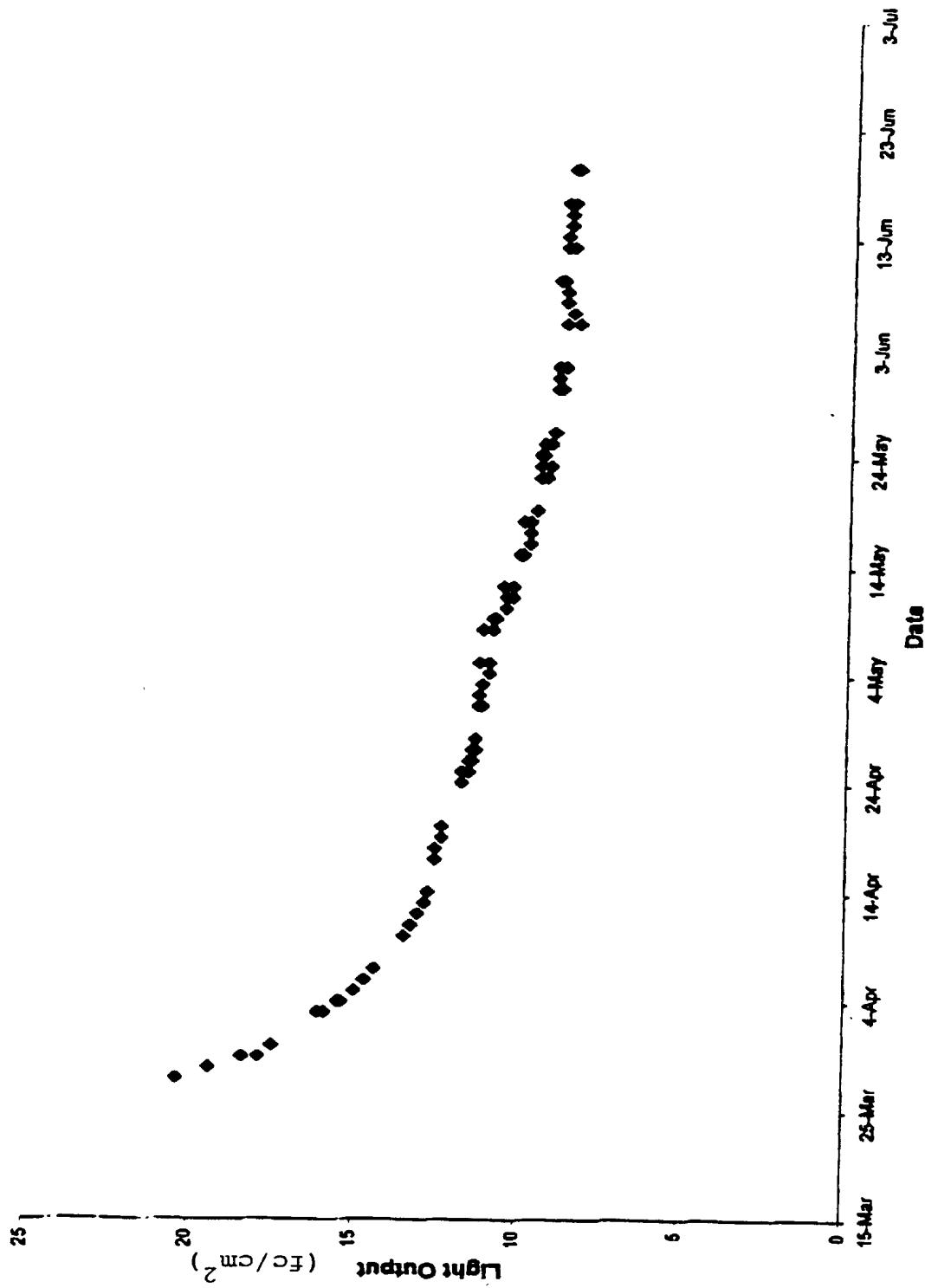
(57) Abstract: The present invention discloses an ultraviolet light curable electroluminescent composition and method for making such a composition that may be used to produce an electroluminescent active layer. An active layer is a layer that when incorporated in a suitable device emits light when a voltage is applied. The disclosed composition does not contain any significant amount of volatile organic solvents that do not become incorporated in the active layer after curing. It is an advantage of the present invention that the deposition of the dielectric layer in such a device is optional when the disclosed electroluminescent composition is used to deposit the active layer.

WO 01/25342 A1

10-089,960

1/1

FIGURE 1



DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

Atty. Docket No. KRO0105PUSA
First Named Inventor Roy C. Krohn

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

UV CURABLE COMPOSITIONS FOR PRODUCING ELECTROLUMINESCENT COATINGS

the specification of which:

is attached hereto; or
 was filed on (MM/DD/YYYY) October 5, 2000 as U.S. Application Number or **PCT International Application Number** PCT/US00/41086, and was amended on (MM/DD/YYYY) _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below, and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Priority Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? (Yes/No)
.....

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)
60/157,944	October 6, 1999

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, § 1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application.

Application Number(s)	Filing Date (MM/DD/YYYY)	Status: Patented, Pending, Abandoned
.....

Declaration for Patent Application (cont'd.)Atty. Docket No. KRO0105PUSA

I hereby appoint the following registered practitioners to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

Ernie L. Brooks, Reg No 26,260, James A. Kushman, Reg No 25,634, David R. Syrowik, Reg No 27,956, Mark A. Cantor, Reg No 30,614, Ralph M. Burton, Reg No 17,748, Robert C.J. Tuttle, Reg. No. 27,962, Earl J. LaFontaine, Reg. No. 30,766, Ronald M. Nabozny, Reg. No. 28,648, Thomas A. Lewry, Reg. No. 30,770, John E. Nemazi, Reg. No. 30,826, Kevin J. Heml, Reg. No. 29,805, William G. Abbott, Reg. No. 31,936; Donald J. Harrington, Reg. No. 17,427, Frederick M. Ritchie, Reg. No. 18,669; Robert C. Brandenburg, Reg. No. 29,048, A. Frank Duke, Reg. No. 20,937; John M. Halan, Reg. No. 35,534, Jeffrey M. Szuma, Reg. No. 35,700, James R. Ignatowski, Reg. No. 26,741, Frank A. Angileri, Reg. No. 36,733, William G. Conger, Reg. No. 31,209, Sangeeta G. Shahi, Reg. No. 38,614, Christopher W. Quinn, Reg. No. 38,274, Robert C. Jones, Reg. No. 35,209, David S. Bir, Reg. No. 38,383, James N. Kallis, Reg. No. 41,102, Hugo A. Delevie, Reg. No. 32,688; Michael S. Brodbine, Reg. No. 38,392, Jeremy J. Cercuui, Reg. No. 42,454, Mark D. Chuey, Reg. No. 42,415, Pete N. Kiousis, Reg. No. 41,117, Stephanie M. Mansfield, Reg. No. 43,773, Mark E. Stuenkel, Reg. No. 44,364, Matthew R. Mowers, Reg. No. 44,956, Ginta Kukainis, Reg. No. 46,082; Seth E. Rodack, Reg. No. 45,622, Matthew M. Jakubowski, Reg. No. 44,801, Marc Lorelli, Reg. No. 43,759, John S. Le Roy, Reg. No. 48,158; Thomas W. Cunningham, Reg. No. 48,722, John D. Russell, Reg. No. 47,048, Marc F. Malooley, Reg. No. P-50,624.

Address all correspondence and telephone calls to Michael S. Brodbine
at Brooks & Kushman P.C., 1000 Town Center, Twenty-Second Floor, Southfield, Michigan 48075, (248) 358-4400.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

100 **Full Name of Sole or First Inventor**

Roy C. KrohnInventor's signature *R.C. Krohn*Date 3/14/02Post Office Address 3540 Orvall Drive, Fort Gratiot, Michigan 48059Residence (as above)Citizenship U.S.A.MI**Full Name of Second Joint Inventor**

Inventor's signature

Date

Post Office Address

Residence

Citizenship

Full Name of Third Joint Inventor

Inventor's signature

Date

Post Office Address

Residence

Citizenship

Full Name of Fourth Joint Inventor

Inventor's signature

Date

Post Office Address

Residence

Citizenship